Review of the ACP manuscript acp-2009-553

"Particle formation in the Arctic free troposphere during the ASTAR 2004 campaign: a case study on the influence of vertical motion on the binary homogeneous nucleation of H_2SO_4/H_2O"

by Khosrawi et al.

General Comments:

The manuscript "Particle formation in the Arctic free troposphere during the ASTAR 2004 campaign: a case study on the influence of vertical motion on the binary homogeneous nucleation of H_2SO_4/H_2O" by Khosrawi et al. deals with the interpretation of one particular particle formation event in the Arctic free troposphere. The authors use in-situ measured, aircraft-borne data, backward trajectories, and a microphysical box model to investigate under which conditions particle formation occurs.

Unfortunately, I have to say that the manuscript contains many small errors and inaccuracies, which distract the reader from the scientific contents. I have the feeling that it was written too hasty and not checked properly before submission.

Concerning the contents, I have to admit that I have two problem with the manuscript. The first is concerning the relevance of the results. Particle number concentration measurements in the Artic troposphere are rare and hence represent valuable data, however this study does not focus on the data, but on nucleation process modeling. This nucleation process is investigated with a known methodology. The result of the analysis is that the nucleation depends on the type of vertical upwards motion, slow or fast. But such a dependency on the motion of the air mass could be expected, or? What I´m missing is that the authors convince me as a reader, why such a study is of relevance to atmospheric science.

The second problem I see is with the line of arguments. The model indicates particle formation for all investigated trajectories, which makes me doubt if it is the appropriate tool to investigate why nucleation mode particles were found on one day, but not on the other two. Furthermore, differences in air mass history is not visible, because there is no trace gas information available, and the shown trajectories do not include altitude information. The authors consider the three days (within a period of five days) to be comparable concerning chemical composition and atmospheric conditions, only upward motion seems to be different. Again, the authors might be correct, but please convince the reader that not different air mass composition instead of the vertical motion is the main reason for the observed differences in particle number density.

If the authors can improve the manuscript with respect to these two central questions, and correct all the small errors and inaccuracies, the manuscript might become a valuable reference.

Specific Comments:

- p. 21961, Introduction, first paragraph: the link between the climate issue and particle formation is missing, why is particle formation important, and why in the Arctic?
- p. 21961, Introduction, second paragraph: the authors present the different particle formation processes discussed in the literature, but this presentation does not include the recent papers to this topic, e.g., Laaksonen et a., ACP 8, 7255-7264, 2008. Furthermore, they should argue more clearly, why it is valid to use the binary homogeneous nucleation for modeling particle formation in the free troposphere (cf. e.g., introduction in Laaksonen et al, 2008, Weber et al., 1999).
- p. 21962 l. 1, the Hermann et al., 2003 reference is poorly cited. These authors did not measure the particle size distribution, but only integral particle number concentrations in two size ranges, or? Furthermore, they found that atmospheric dynamics (in this case deep convective clouds) also play an essential role in the tropics, not only at mid-latitudes.
- p. 21962 l. 6, particle formation due to stratosphere-troposphere exchange was also found by Zahn, A. et al., J. Geophys. Res. 105, 1527-1535, 2000, please add this reference.
- p. 21964 l. 3 …, please state exactly which instruments you were using, i.e. the manufacturer and the model types.
- p. 21964 l. 8, the authors emphasize the uncertainties associated with counting statistics, but what about the uncertainties caused by the particle sampling efficiency or the inlet system, pressure dependent counting efficiencies, etc. Please add this important information.
- p. 21964 l. 17, the stated uncertainty of the FSSP-300 refers to what, sizing or concentration? Other investigators give higher numbers, e.g., http://www.eol.ucar.edu/raf/Bulletins/B24/fssp300.html.
- p. 21964 l. 21, in terms of the above statement, I don't believe the 5 % accuracy of the CPSA to be true, it should be higher.
- p. 21965 l. 1, the estimation about the particle losses is based on what? And if 35% of the 4 nm particles get lost in the sampling system (on average maybe 20% for the size range 4-10 nm) this number is large and the data should be corrected for using an average value, even without the knowledge of the exact size distribution.
- p. 21965 l. 4, are there any trace gas measurements available, which might help interpreting the data?
- p. 21965 l. 11, please add a figure showing the flight tracks.
- p. 21965 l. 11, it might be nitpicking, but personally I feel that the first figure mentioned in the text should also get the figure number "1", and the order of figures should not be mixed.
- p. 21966 l. 2, I'm not a specialist in modeling particle nucleation, but aren't there more recent, improved formulations for the homogeneous nucleation theory, e.g., F. Yu, J. Chem. Phys. 127, 054301, 2007?

- p. 21966 l. 5, condensation of material leads to changes in particles size, which influences the particle coagulation rate. So, if you do not consider condensation, how larger is the uncertainty in your modeling due to this neglect?
- p. 21966 l. 7, please provide a reference for the statement in the last sentence of this paragraph.
- p. 21966 l. 27, which “three” trajectories are meant? Either there are four (2x 24 May, 1x 22 and 26 May, respectively) or six, if there were trajectories for both altitudes for each of the three days.
- p. 21967 l. 4, sorry, but I could not follow your counting, how did you come to 36 trajectories in total? Please explain in a way that the reader understands why you chose this number.
- p. 21967 l. 12, why do you need the stability parameters to know if vertical motion has occurred along the trajectory? What kind of additional information do they provide, which is not provided by the trajectory altitude or pressure?
- p. 21968 l. 5, did I understand it right that the box model calculations were initialized with measurements made six days later? If, yes, please provide some evidence that this approach is valid.
- p. 21968 l. 28, please indicate that the sulfuric acid concentrations are gas phase concentrations. The chosen values are high, according to your references typical for polluted air masses. Why did you chose these high values, the trajectories suggest rather clean air masses, or?
- p. 21969 l. 9, in line 6 you used the order 40 pptv, 80 pptv, and 1 pptv for the sulfuric acid mixing ratio, hence in line 9 one would assume that the nucleation time steps 17 h, 416 h, and 2 h are for the same order of sulfuric acid mixing ratios, which is not the case, or?
- p. 21969 l. 15, I do not fully understand the differentiation between temperature and vertical motion as trigger for new particle formation. Vertical motion (disregarding the reduction of particle surface area due to scavenging processes inside clouds and different photochemistry at higher altitudes) is equivalent to temperature change, so why make a difference? Which process, besides altitude change should be responsible for the temperature change? Please explain.
- p. 21969 l. 26, as figures 4 to 6 show, the model indicates particle formation along all 36 trajectories. Is this realistic? I would assume that there are trajectories which are not accompanied by particle nucleation. Otherwise, it would mean that in the region under investigation particle formation is always taking place.
- p. 21969 l. 28, the gamma values are what? Mean values along the trajectory, stability at the time when the maximum particle concentration occurred, maximum stability along the trajectory, …?
- p. 21970 the two paragraphs comparing the three cases in Fig. 4-6: I read the paragraphs two times, but I did not get the message (sorry, maybe I´m too tired). What do you want to show

with figures? How is your “uniformity” defined (for the 1 pptv data in Fig. 4 the number densities vary over nearly three orders of magnitude)? And why does the “uniformity agree with the behavior found for case 1”? What “relationship” are you talking about?

- p. 21970 l. 13, is it justified to declare a “slight decrease” in the number density based on only two data points. I don´t think so.
- p. 21970 l. 26, if you discuss the relation between the number density and the updraft velocity displayed in Fig. 7 in only one sentence, is it necessary to show this figure?
- p. 21971 l. 16, the 24 May is not “dominated” by case 1, because case 2 has nearly the same number of occurrence. Please change this statement.
- p. 21972 l. 5, which were the criteria by which the trajectories were chosen for Fig. 8, i.e., why there are some trajectories included and others not?
- p. 21972 l. 5, what do you mean by “… some trajectories … lie also within this area”? Do you refer to a geographical area?
- p. 21973 l. 1, if on 26 May 1 pptv sulfuric acid leads to a too low nucleation mode particle number density and 40 pptv as well as 80 pptv lead to too strong nucleation events (and thereby again to a low nucleation mode particle number density at the measurement time), a sulfuric acid mixing ratio “somewhat lower than 40 pptv” should lead to a nucleation mode particle number density which should haven been observable, or?

Technical Comments:

- again, it seems to me that the manuscript was written hastily, because there are several simple language errors, e.g., p. 21960, l. 6, “vertical motion are the reason”, p. 21963 l. 17, “aircrafts”, p. 21967, l. 1, “coordinates which represents”. Please check the whole document again.
- p. 21961 l. 10, I never heard the term “nucleation … conversion”, and I believe it to be wrong. It should be either “particle nucleation”, “nucleation process”, or “gas-to-particle conversion”, but not a mixture of these two terms (which are not the same, by the way).
- p. 21962 l. 8, please insert “particle” before “properties”.
- p. 21964 l. 7, “CPC” must be “OPC”.
- p. 21967 l. 1, please use a consistent nomenclature for the geographical coordinates, i.e., either always separated by a slash or never.
- p. 21969 l. 13, the trajectory start should be at 19.0°E and 78.7°N, or is it a new trajectory? Please see also Fig. 3.

- p. 21969 l. 18, please remove “previously” because 1999 (and there are other references as well) is ten years ago.
- p. 21969 l. 20, the reference should be to Table 2, not Table 1.
- p. 21971 l. 3, please remove the “and” before “to case 3”.

- Tab 1: “uK/Pa” should be “µK/Pa”, or? Same in Tab. 3.
- Tab 3: in the table “Altitude” without “d”.
- Fig. 1: the trajectories do not start at 10° or 11°E, they all start at 22.5°E!
- Fig. 1: please include trajectory altitude information in the figure, e.g., by using different shades of color.
- Fig. 2: the chosen representation of data wastes 20% of the figure, because they are not used, why do you not expand the x-axis scale, which would make the structures more visible?
- Fig. 2: there are gaps in the N_4 time series, in particular for the flight altitude 7000 m, please explain and add this information to the text. What are the implications for your simulation?
- Fig. 2, legend: “begin” should be “beginning”, or?
- Fig. 2, legend: the reference to the simulation is a little bit confusing, because one might think that the data were simulated.
- Fig. 4: symbols are hard to see, please use solid symbols. Same for Fig. 5, 6, and 7.
- Fig. 4, x-axis: “uK” should be “µK”, or? Same for Fig. 5 and 6.
- Fig. 5, upper, right graph: there seems to be one green and one blue symbol missing, or do they fall together with two other data points? If yes, please notify this.